Optical scattering by a nonlinear medium, I: from Maxwell's equations to numerically tractable equations

Pierre Godard, Frédéric Zolla, André Nicolet

Institut Fresnel UMR CNRS 6133, Faculté de Saint-Jérôme case 162

13397 Marseille Cedex 20, France

pierre.godard@fresnel.fr,frederic.zolla@fresnel.fr,andre.nicolet@fresnel.fr

Abstract

A new method to find the propagation equation system governing the scattering of an electromagnetic wave by a nonlinear medium is proposed. The aim is to let the effects appear spontaneously, deleting as far as possible the phenomenological ideas. In this way, we obtain propagation equation systems that encodes several nonlinear effects. Once these systems obtained, the numerical values of the tensors that characterize the answer of the medium to an electromagnetic perturbation give weights to the different effects.

This aim is partly reached in this study, especially when treating harmonic generations. For this, we start from the Maxwell's equation and give rigorously all the hypothesis needed to attain equation systems that can be solved, at least from a numerical point of view. Finally, a symmetry on the susceptibility tensors that ensures that a medium is lossless from the electromagnetic point of view is worked out.

1 Introduction

The usual route to treat nonlinear media from the electromagnetic point of view is to expand the electric permittivity as a function of the electric field. These expansions quickly get huge and so one has first to select the effect we want to consider, and then systematically simplify the equations, keeping only the terms that contribute to this effect ([1, 2]). A look at the literature shows that the precision that can be obtained with this method is out of question.

Nevertheless, it would appear more satisfactory to start only with Maxwell's equations and to let the effects appear by themselves, introducing a systematical way to simplify the equations. The simplification is thus *a priori*, generic ; the relative importance of the observed effects being only *a posteriori* determined, by the numerical values of the susceptibility tensors. This paper aims at drawing this new road.

For this, some general work on the constitutive relations had to be done. We expose it in the subsection 2.1. Then, with some assumptions, we recover in the next subsection the standard expression of the electric susceptibility tensors. Most of these assumptions are justified for the presentation of this paper, and avoiding them would not have lead to insurmountable difficulties.

Having expressed the answer of a medium, we obtain the propagation equation system satisfied by the electric field in 3.1. This system, parameterized by a continuous parameter, is far too complicated to be solved, even with numerical methods. This leads us to partly leave our general aim, and concentrate on harmonic generation. In this context, we introduce a new notion, called the degree: it determines, in generic terms, which interaction between the Fourier components of the electric field have to be taken into account. The subsections 3.3 and 3.4 are devoted to the propagation equation systems in the lowest order of nonlinearity and in the lowest degree; the physical effects are then clearly identified. We hope the obtention of these equation systems from *ab initio* principles will convince the reader about the interest of the method.

In a second part (section 4), we try to answer, we think more rigorously that what can be found in the literature, to the question: `how the electric energy variation is determined by the susceptibility tensor fields of a medium?' If the general answer is still unknown, we give a sufficient condition for a medium to be lossless.

This paper is intended to be appreciated by the theoretical physicists, either working in the nonlinear optics field or not, as well as the applied physicists. To this aim, we start from general concepts and end with equation systems that can be numerically solved. Having clarified the way these systems appear, we have realized some simulations, one of them appearing in the companion paper [3]. In particular, these simulations confirmed our prediction on the energy criterion.

2 The electromagnetic constitutive relations

2.1 A tractable expression for the inductive fields

As said in the introduction, the aim of this article is to expose a new route to the equations of nonlinear optics, starting only with the Maxwell's equations. We will work at a completely classical level, even if, of course, it is hoped that the effective characteristics of the medium will in future works clearly come from a microscopic point of view. In other words, in this study, the characterization of a medium is considered to be fully determined as soon as a set of (susceptibility) tensors are given.

Let us start with the Maxwell's equations. We consider here that the primitive fields are the electric field $\mathbf{E}$ and the magnetic field $\mathbf{B}$. The first set of equations gives a 'coherence' condition between $\mathbf{E}$ and $\mathbf{B}$:

$$\begin{pmatrix} \nabla\times\mathbf{E}+\partial_t\mathbf{B} \\ \nabla\cdot\mathbf{B} \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \end{pmatrix}. \tag{1}$$

The second set of equations links the induction fields with the sources of the electromagnetic fields. We thus denote by $\mathbf{D}$ the electric induction, $\mathbf{B}$ the magnetic induction,

ρ the charge density and $\mathbf{J}$ the current density. Then, the second set of Maxwell's equations is

$$\begin{pmatrix} \nabla \times \mathbf{H} - \partial_t \mathbf{D} \\ \nabla \cdot \mathbf{D} \end{pmatrix} = \begin{pmatrix} \mathbf{J} \\ \rho \end{pmatrix}. \tag{2}$$

Supposing that the sources are known, we are looking for the electromagnetic fields. It appears that we have to add some hypothesis to answer this problem, for, in each point $\mathbf{s}$ of $\mathbb{R}^3$ and for any time t, we have twelve unknowns (the three components of the four vectors $\mathbf{E}(\mathbf{s},t)$, $\mathbf{B}(\mathbf{s},t)$, $\mathbf{D}(\mathbf{s},t)$ and $\mathbf{H}(\mathbf{s},t)$), but the Maxwell's equations give only two vectorial and two scalar equalities. The set of Maxwell's equations being independent of the medium in which the fields oscillate, we have to encode the (electromagnetic) characteristics of the medium, with two (vectorial) relations that give the inductive fields in function of the primitive fields. We pose the following set of equations:

$$\begin{cases} \mathbf{D}(\mathbf{s},t) = \mathfrak{D}\big(\{\mathbf{E}(\boldsymbol{\sigma},\tau)\}_{(\boldsymbol{\sigma},\tau)\in S_e^d(\mathbf{s})\times T_e^d(t)}, \{\mathbf{B}(\boldsymbol{\sigma},\tau)\}_{(\boldsymbol{\sigma},\tau)\in S_b^d(\mathbf{s})\times T_b^d(t)}, \mathbf{s}, t\big) & (3) \\ \mathbf{H}(\mathbf{s},t) = \mathfrak{H}\big(\{\mathbf{E}(\boldsymbol{\sigma},\tau)\}_{(\boldsymbol{\sigma},\tau)\in S_e^h(\mathbf{s})\times T_e^h(t)}, \{\mathbf{B}(\boldsymbol{\sigma},\tau)\}_{(\boldsymbol{\sigma},\tau)\in S_b^h(\mathbf{s})\times T_b^h(t)}, \mathbf{s}, t\big) & (4) \end{cases}$$

where, for example, $\{\mathbf{E}(\boldsymbol{\sigma},\tau)\}_{(\boldsymbol{\sigma},\tau)\in S_e^d(\mathbf{s})\times T_e^d(t)}$ is the set of all values of the electric field at the place $\boldsymbol{\sigma}$ and time τ, when $\boldsymbol{\sigma}$ and τ run over the sets $S_e^d(\mathbf{s}) \subset \mathbb{R}^3$ and $T_e^d(t) \subset \mathbb{R}$ respectively. As said above, the 'fabrication' of the electromagnetic induction is done at another scale than the one considered here, and we are reduced to see it like a black box: at the input are, for each place $\mathbf{s}$ and each time t, the primitive fields, and $\mathbf{s}$ and t; at the output are the inductive vectors $\mathbf{D}(\mathbf{s},t)$ and $\mathbf{H}(\mathbf{s},t)$. Several authors ([4, 5], see also [6]) give expressions similar to (3-4), but we think that the one we present is more rigorous and allows a clear transcription of the properties of the medium. The article [7], and some references found here, is not restricted to pure electromagnetic phenomena.

The transcriptions, between the properties of the medium and the restriction it induces on the functionals $\mathfrak{D}$ and $\mathfrak{H}$ are the following one:

- The causality of the medium implies that

$$T_a^b(t) \subset (-\infty, t], \quad (a,b) \in \{e,b\} \times \{d,h\},$$

  which means that the times τ at which are evaluated the electric and magnetic fields that contribute to $\mathbf{D}(\mathbf{s},t)$ and $\mathbf{H}(\mathbf{s},t)$ are prior to t.

- The locality (in space) of the medium implies that

$$S_a^b(\mathbf{s}) = \{\mathbf{s}\}, \quad (a,b) \in \{e,b\} \times \{d,h\}.$$

  Allowing the notations' abuse that consists in keeping the same names for the inductive functionals, the new expression are

$$\left\{\begin{array}{l}\mathbf{D}(\mathbf{s},t)=\mathfrak{D}\big(\{\mathbf{E}(\mathbf{s},\tau)\}_{\tau\in T_e^d(t)},\{\mathbf{B}(\mathbf{s},\tau)\}_{\tau\in T_b^d(t)},\mathbf{s},t\big)\\ \mathbf{H}(\mathbf{s},t)=\mathfrak{H}\big(\{\mathbf{E}(\mathbf{s},\tau)\}_{\tau\in T_e^h(t)},\{\mathbf{B}(\mathbf{s},\tau)\}_{\tau\in T_b^h(t)},\mathbf{s},t\big).\end{array}\right.$$

- A medium is nonbianisotropic if

$$\left\{\begin{array}{l}\partial_{\mathbf{B}}\mathfrak{D}=0\\ \partial_{\mathbf{E}}\mathfrak{H}=0.\end{array}\right.$$

  In this case, we have

$$\left\{\begin{array}{l}\mathbf{D}(\mathbf{s},t)=\mathfrak{D}\big(\{\mathbf{E}(\boldsymbol{\sigma},\tau)\}_{(\boldsymbol{\sigma},\tau)\in S_e^d(\mathbf{s})\times T_e^d(t)},\mathbf{s},t\big)\\ \mathbf{H}(\mathbf{s},t)=\mathfrak{H}\big(\{\mathbf{B}(\boldsymbol{\sigma},\tau)\}_{(\boldsymbol{\sigma},\tau)\in S_b^h(\mathbf{s})\times T_b^h(t)},\mathbf{s},t\big)\end{array}\right.$$

- A medium is homogeneous in time, or, as is commonly said, stationary, if the functionals do not explicitly depend on the time t:

$$\left\{\begin{array}{l}\partial_t\mathfrak{D}=0\\ \partial_t\mathfrak{H}=0.\end{array}\right.$$

  The constitutive relations then get:

$$\left\{\begin{array}{l}\mathbf{D}(\mathbf{s},t)=\mathfrak{D}\big(\{\mathbf{E}(\boldsymbol{\sigma},\tau)\}_{(\boldsymbol{\sigma},\tau)\in S_e^d(\mathbf{s})\times T_e^d(t)},\{\mathbf{B}(\boldsymbol{\sigma},\tau)\}_{(\boldsymbol{\sigma},\tau)\in S_b^d(\mathbf{s})\times T_b^d(t)},\mathbf{s}\big)\\ \mathbf{H}(\mathbf{s},t)=\mathfrak{H}\big(\{\mathbf{E}(\boldsymbol{\sigma},\tau)\}_{(\boldsymbol{\sigma},\tau)\in S_e^h(\mathbf{s})\times T_e^h(t)},\{\mathbf{B}(\boldsymbol{\sigma},\tau)\}_{(\boldsymbol{\sigma},\tau)\in S_b^h(\mathbf{s})\times T_b^h(t)},\mathbf{s}\big).\end{array}\right.$$

  We would like to stress that this hypothesis is, in a way, delicate, when treating nonlinear optics. Indeed, for the nonlinear effects to be important, the intensity of light has to be large. But if this intensity is too large, then we cannot neglect the effects of the electromagnetic field on the medium. Some effects like saturation, damage, etc. do appear, and we leave the realm of stationary media.

The transcriptions of locality in time (commonly called nondispersive) or homogeneity in space in the inductive functions should be obvious. We can then define the (local) electromagnetic vacuum: a vacuum holds at a point $\mathbf{s}$ and a time t if the following equalities of vectors are satisfied:

$$\left\{\begin{array}{l}\mathbf{D}(\mathbf{s},t)=\varepsilon_0\,\mathbf{E}(\mathbf{s},t)\\ \mathbf{H}(\mathbf{s},t)=1/\mu_0\,\mathbf{B}(\mathbf{s},t).\end{array}\right.$$

This leads to define the ‘answer’ of a medium to an electromagnetic perturbation as the difference between the induction in that medium and the one in a vacuum:

$$\left\{\begin{array}{l} \mathbf{P}_e := \mathbf{D} - \varepsilon_0\,\mathbf{E} \\ \mathbf{P}_m := \mathbf{H} - 1/\mu_0\,\mathbf{B}. \end{array}\right.$$

These two vector fields are called the electric polarization vector field and the magnetic polarization vector field - we prefer to write $\mathbf{P}_m$ for what is usually denoted by $\mathbf{M}$ (termed the magnetization vector field) for the coherence of this article. We now arrive at a key step. In the following definition, we denote by V' the dual of the vector space V.

Definition 1 *Smooth Medium*

A nonbianisotropic medium is called smooth in a neighborhood of a point $\mathbf{s}$ *and a time* t *if the electric and magnetic polarization vector fields admit Taylor expansions of the following kind:*

$$\left\{\begin{array}{l} \mathbf{P}_e = \mathbf{P}_e^{(0)} + \sum\limits_{n\in\mathbb{N}} \mathbf{P}_e^{(n)} \\ \mathbf{P}_m = \mathbf{P}_m^{(0)} + \sum\limits_{n\in\mathbb{N}} \mathbf{P}_m^{(n)} \end{array}\right.$$

with

$$\left\{\begin{array}{l} \mathbf{P}_e^{(n)}(\mathbf{s},t) = \varepsilon_0 \displaystyle\int_{-\infty}^{\infty} dt_1 \cdots \int_{-\infty}^{\infty} dt_n\, Q_e^{(n)}(\mathbf{s},t;\mathbf{s}_1,t_1,\cdots,\mathbf{s}_n,t_n) \\ \qquad \mathbf{E}(\mathbf{s}_1,t_1)\cdots\mathbf{E}(\mathbf{s}_n,t_n) \\ \mathbf{P}_m^{(n)}(\mathbf{s},t) = \varepsilon_0 \displaystyle\int_{-\infty}^{\infty} dt_1 \cdots \int_{-\infty}^{\infty} dt_n\, Q_m^{(n)}(\mathbf{s},t;\mathbf{s}_1,t_1,\cdots,\mathbf{s}_n,t_n) \\ \qquad \mathbf{H}(\mathbf{s}_1,t_1)\cdots\mathbf{H}(\mathbf{s}_n,t_n) \end{array}\right.$$

where

$$\left\{\begin{array}{l} Q_e^{(n)} : \mathbb{R}^4 \times \mathbb{R}^{4n} \to \mathbb{R}^3 \otimes (\mathbb{R}^3)'^{\otimes n} \\ Q_m^{(n)} : \mathbb{R}^4 \times \mathbb{R}^{4n} \to \mathbb{R}^3 \otimes (\mathbb{R}^3)'^{\otimes n}. \end{array}\right.$$

A similar definition can be applied to bianisotropic media as well: the tensors $Q_e^{(n)}$ and $Q_m^{(n)}$ have then to be contracted with the 2^n combinations of $\mathbf{E}$ and $\mathbf{B}$. Though treating these media, among which we find the chiral ones, do not bring important new difficulties, we won't anymore be concerned with them, to keep formulae of 'reasonable' size.

Due to the nonbianisotropy hypothesis, the electric and magnetic inductions can be treated separately. We will from now on concentrate on the electric one. Hence, when no confusion is possible, we will write $\mathbf{P}$ in place of $\mathbf{P}_e$, $Q^{(n)}$ is place of $Q_e^{(n)}$, say

the polarization vector instead of the electric polarization vector, etc. The treatment required for the magnetic part is completely similar.

The set of operators $Q^{(n)}$, when applied to $(\mathbf{s}, t; \mathbf{s}_1, t_1, \cdots, \mathbf{s}_n, t_n)$, give the effect, at the point $\mathbf{s}$ and time t, that the electric field, evaluated at the points $\mathbf{s}_1, \cdots, \mathbf{s}_n$ and times $t_1, \cdots, t_n$ have on the electric polarization vector at the point $\mathbf{s}$ and time t. The zero-th order of the electric polarization vector $\mathbf{P}^{(0)}(\mathbf{s}, t)$ corresponds to a spontaneous nonzero electric moment. It does not vanish for ferroelectric materials. For convenience, we write $\mathbf{P}^{(0)}(\mathbf{s}, t) = \varepsilon_0 Q^{(0)}(\mathbf{s}, t)$. The term $\mathbf{P}^{(n)}$, for $n \in \mathbb{N}$ is called the n-th order of the polarization vector.

2.2 A tractable expression for the polarization vector

2.2.1 The definition of the electric susceptibility tensors

The expansion of the (electric) polarization vector given in the definition (1) is still too complicated, and needs further simplification to be studied. The following hypothesis, still motivated for the presentation, is that the medium is spatially local. The electric inductive functional has the form

$$\mathbf{D}(\mathbf{s}, t) = \mathfrak{D}\big(\{\mathbf{E}(\mathbf{s}, \tau)\}_{\tau \in T_e^d(t)}, \mathbf{s}, t\big)$$

so that

$$\begin{aligned}\mathbf{P}(\mathbf{s}, t) = \mathbf{P}^{(0)}(\mathbf{s}, t) + \sum_{n \in \mathbb{N}} \varepsilon_0 \int_{-\infty}^{\infty} dt_1 \cdots \int_{-\infty}^{\infty} dt_n \, Q^{(n)}(\mathbf{s}, t; t_1, \cdots, t_n) \\ \mathbf{E}(\mathbf{s}, t_1) \cdots \mathbf{E}(\mathbf{s}, t_n). \qquad (5)\end{aligned}$$

Once again, we have adapted the functionals without changing the name (now, we have $Q^{(n)} : \mathbb{R}^4 \times \mathbb{R}^n \to \mathbb{R}^3 \otimes (\mathbb{R}^3)'^{\otimes n}$).

Of more fundamental nature, let us suppose that the medium is stationary; this means that, first, the spontaneous polarization does not depend on time, and secondly, that for any point $\mathbf{s}$ and for any duration time T, the effect of the electric vector field evaluated at the point $\mathbf{s}$ and at the times $t_1, \cdots, t_n$ on the polarization vector field evaluated at the point $\mathbf{s}$ and at the time t is the same than the effect of the electric vector field evaluated at the point $\mathbf{s}$ and at the times $t_1 - T, \cdots, t_n - T$ on the polarization vector field evaluated at the point $\mathbf{s}$ and at the time $t - T$, i.e.

$$Q^{(0)}(\mathbf{s}, t) = Q^{(0)}(\mathbf{s}, t - T)$$

and

$$Q^{(n)}(\mathbf{s}, t; t_1, \cdots, t_n) = Q^{(n)}(\mathbf{s}, t - T; t_1 - T, \cdots, t_n - T), \quad \forall n \in \mathbb{N}, \forall T \in \mathbb{R}.$$

When considering stationary media, a usual trick ([2]) is to define

$$\begin{array}{rccl} R^{(0)} : & \mathbb{R}^3 & \to & \mathbb{R}^3, \\ & \mathbf{s} & \mapsto & Q^{(0)}(\mathbf{s}, 0), \end{array}$$

$$\begin{array}{rlcl} R^{(n)}: & \mathbb{R}^3 \times \mathbb{R}^n & \to & \mathbb{R}^3 \otimes (\mathbb{R}^3)'^{\otimes n}, \\ & (\mathbf{s}, t_1, \cdots, t_n) & \mapsto & Q^{(n)}(\mathbf{s}, 0; -t_1, \cdots, -t_n), \quad n \in \mathbb{N}. \end{array}$$

This function $R^{(n)}$ is called the response function of the n-th order, because it allows to write the relation (5) as a convolution on $\mathbb{R}^n$ between $R^{(n)}$ and

$$\mathbf{E}^{(n)} : \mathbb{R}^3 \times \mathbb{R}^n \to (\mathbb{R}^3)^{\otimes n}, (\mathbf{s}, t_1, \cdots, t_n) \mapsto \mathbf{E}(\mathbf{s}, t_1) \cdots \mathbf{E}(\mathbf{s}, t_n) \; :$$

$$\begin{aligned} \mathbf{P}^{(n)}(\mathbf{s}, t) &= \varepsilon_0 \big(R^{(n)}(\mathbf{s}, \cdot) *_n \mathbf{E}^{(n)}(\mathbf{s}, \cdot) \big)(t) \\ &= \varepsilon_0 \int_{-\infty}^{\infty} dt_1 \cdots \int_{-\infty}^{\infty} dt_n \, R^{(n)}(\mathbf{s}, t - t_1, \cdots, t - t_n) \mathbf{E}(\mathbf{s}, t_1) \cdots \mathbf{E}(\mathbf{s}, t_n) \end{aligned}$$

In the last equation, we make the transformation $t_j \mapsto t - t_j$ to obtain

$$\mathbf{P}^{(n)}(\mathbf{s}, t) = \varepsilon_0 \int_{-\infty}^{\infty} dt_1 \cdots \int_{-\infty}^{\infty} dt_n \, R^{(n)}(\mathbf{s}, t_1, \cdots, t_n) \mathbf{E}(\mathbf{s}, t - t_1) \cdots \mathbf{E}(\mathbf{s}, t - t_n)$$

and then we express the electric field in its Fourier basis,

$$\mathbf{E}(\mathbf{s}, t - t_j) = \int_{-\infty}^{\infty} d\omega_j \, e^{-i\omega_j (t - t_j)} \hat{\mathbf{E}}(\mathbf{s}, \omega_j), \quad j \in \{1, \cdots, n\}.$$

This leads to

$$\begin{aligned} \mathbf{P}^{(n)}(\mathbf{s}, t) &= \varepsilon_0 \int_{-\infty}^{\infty} d\omega_1 \, e^{-i\omega_1 t} \cdots \int_{-\infty}^{\infty} d\omega_n \, e^{-i\omega_n t} \\ & \qquad \underline{\chi}^{(n)}(\mathbf{s}, \omega_1, \cdots, \omega_n) \hat{\mathbf{E}}(\mathbf{s}, \omega_1) \cdots \hat{\mathbf{E}}(\mathbf{s}, \omega_n) \\ &= \varepsilon_0 \int_{-\infty}^{\infty} d\omega_1 \cdots \int_{-\infty}^{\infty} d\omega_n \, \underline{\chi}^{(n)}(\mathbf{s}, \omega_1, \cdots, \omega_n) \\ & \qquad \hat{\mathbf{E}}(\mathbf{s}, \omega_1) \cdots \hat{\mathbf{E}}(\mathbf{s}, \omega_n) e^{-i(\omega_1 + \cdots + \omega_n) t} \end{aligned} \tag{6}$$

where

$$\begin{array}{rcl} \underline{\chi}^{(n)} : \mathbb{R}^3 \times \mathbb{R}^n & \to & \mathbb{C}^3 \otimes (\mathbb{C}^3)'^{\otimes n}, \\ (\mathbf{s}, \omega_1, \cdots, \omega_n) & \mapsto & \displaystyle\int_{-\infty}^{\infty} dt_1 \cdots \int_{-\infty}^{\infty} dt_n \, R^{(n)}(\mathbf{s}, t_1, \cdots, t_n) e^{i\boldsymbol{\omega} \cdot \mathbf{t}} \end{array} \tag{7}$$

with $\boldsymbol{\omega} = (\omega_1, \cdots, \omega_n)$, $\mathbf{t} = (t_1, \cdots, t_n)$ and the usual duality product on $\mathbb{R}^n$. We see that $\underline{\chi}^{(n)}$, which is a tensor field of rank $n+1$, called the (electric) susceptibility tensor of the n-th order, is the Fourier transform (up to a scalar multiplication by $(2\pi)^n$) of the response function $R^{(n)}$:

$$\underline{\chi}^{(n)}(\mathbf{s},\omega_1,\cdots,\omega_n) = (2\pi)^n\,\hat{R}^{(n)}(\mathbf{s},\omega_1,\cdots,\omega_n).$$

$\underline{\chi}^{(n)}(\mathbf{s},\omega_1,\cdots,\omega_n)$ gives the way the components, evaluated at the point $\mathbf{s}$, of the electric field oscillating with the angular frequencies ω_1, ..., ω_n contribute to the polarization vector oscillating at the angular frequency $\omega_1+\cdots+\omega_n$ at the same place $\mathbf{s}$. The unit of $\underline{\chi}^{(n)}$ is $m^{n-1}\,V^{1-n}$.

By symmetry, we define $\underline{\chi}^{(0)}$ by

$$\begin{array}{rccl} \underline{\chi}^{(0)}: & \mathbb{R}^3 & \to & \mathbb{R}^3, \\ & \mathbf{s} & \mapsto & R^{(0)}(\mathbf{s}). \end{array}$$

2.2.2 The symmetries of the susceptibility tensors

The susceptibility tensor fields exhibit several symmetries. These ones are important to better understand these tensors, and will allow to greatly simplify the results of the following sections, about propagation equation systems and energy criteria.

The intrinsic permutation symmetry We will now present the intrinsic permutation symmetry of the susceptibility tensors. Although the result of this paragraph is well-known ([1, 2, 8]), we demonstrate it because we think that the way we derive it is more systematic. For this, let us develop the equation (6) in components:

$$P^{(n)\,i}(\mathbf{s},t) = \varepsilon_0 \int_{-\infty}^{\infty} d\omega_1 \cdots \int_{-\infty}^{\infty} d\omega_n\, \underline{\chi}^{(n)\,i}{}_{\;i_1\cdots i_n}(\mathbf{s},\omega_1,\cdots,\omega_n)$$
$$\hat{E}^{i_1}(\mathbf{s},\omega_1)\cdots\hat{E}^{i_n}(\mathbf{s},\omega_n)e^{-i(\omega_1+\cdots+\omega_n)t},$$

where the summation convention over repeated indices is used. The intrinsic permutation symmetry is based on the fact that the $\underline{\chi}^{(n)\,i}{}_{\;i_1\cdots i_n}$ are not uniquely defined: for j in $\{1,\cdots,n\}$, the indexes i_j and the variables ω_j are dummy, so for any bijection σ from $\{1,\cdots,n\}$ to itself, we have:

$$P^{(n)\,i}(\mathbf{s},t) = \varepsilon_0 \int_{-\infty}^{\infty} d\omega_{\sigma 1} \cdots \int_{-\infty}^{\infty} d\omega_{\sigma n}\, \underline{\chi}^{(n)\,i}{}_{\;i_{\sigma 1}\cdots i_{\sigma n}}(\mathbf{s},\omega_{\sigma 1},\cdots,\omega_{\sigma n})$$
$$\hat{E}^{i_{\sigma 1}}(\mathbf{s},\omega_{\sigma 1})\cdots\hat{E}^{i_{\sigma n}}(\mathbf{s},\omega_{\sigma n})e^{-i(\omega_{\sigma 1}+\cdots+\omega_{\sigma n})t}.$$

Thus, denoting by $\mathcal{S}_n$ the symmetric group on a set of cardinality n (we recall that this group is of order $n!$), we have

$$P^{(n)\,i}(\mathbf{s},t) = \frac{\varepsilon_0}{n!}\sum_{\sigma\in\mathcal{S}_n} \int_{-\infty}^{\infty} d\omega_{\sigma 1} \cdots \int_{-\infty}^{\infty} d\omega_{\sigma n}\, \underline{\chi}^{(n)\,i}{}_{\;i_{\sigma 1}\cdots i_{\sigma n}}(\mathbf{s},\omega_{\sigma 1},\cdots,\omega_{\sigma n})$$
$$\hat{E}^{i_{\sigma 1}}(\mathbf{s},\omega_{\sigma 1})\cdots\hat{E}^{i_{\sigma n}}(\mathbf{s},\omega_{\sigma n})e^{-i(\omega_{\sigma 1}+\cdots+\omega_{\sigma n})t}.$$

But, loosely speaking, $\int_{-\infty}^{\infty} d\omega_{\sigma 1} \cdots \int_{-\infty}^{\infty} d\omega_{\sigma n} = \int_{-\infty}^{\infty} d\omega_1 \cdots \int_{-\infty}^{\infty} d\omega_n$ and

$$\prod_{j=1}^{n} \hat{E}^{i_{\sigma j}}(\mathbf{s}, \omega_{\sigma j}) e^{-i\omega_{\sigma j} t} = \prod_{j=1}^{n} \hat{E}^{i_j}(\mathbf{s}, \omega_j) e^{-i\omega_j t}$$

for any σ in $\mathcal{S}_n$. We thus have

$$P^{(n)\,i}(\mathbf{s}, t) = \varepsilon_0 \int_{-\infty}^{\infty} d\omega_1 \cdots \int_{-\infty}^{\infty} d\omega_n \, \{\frac{1}{n!} \sum_{\sigma \in \mathcal{S}_n} \underline{\chi}^{(n)\,i}{}_{i_{\sigma 1} \cdots\, i_{\sigma n}}(\mathbf{s}, \omega_{\sigma 1}, \cdots, \omega_{\sigma n})\}$$
$$\hat{E}^{i_1}(\mathbf{s}, \omega_1) \cdots \hat{E}^{i_n}(\mathbf{s}, \omega_n) e^{-i(\omega_1 + \cdots + \omega_n)t}.$$

This leads to define the (not underlined) tensor $\chi^{(n)} = \mathbf{e}_i \, \chi^{(n)\,i}{}_{i_1 \cdots\, i_n} \otimes \mathbf{e}^{i_1} \otimes \cdots \otimes \mathbf{e}^{i_n}$, once again called susceptibility tensor of order n, by

$$\begin{aligned} \chi^{(n)} : \mathbb{R}^3 \times \mathbb{R}^n \quad &\to \quad \mathbb{C}^3 \otimes (\mathbb{C}^3)'^{\otimes n}, \\ (\mathbf{s}, \omega_1, \cdots, \omega_n) \quad &\mapsto \quad \frac{1}{n!} \sum_{\sigma \in \mathcal{S}_n} \mathbf{e}_i \, \underline{\chi}^{(n)\,i}{}_{i_{\sigma 1} \cdots\, i_{\sigma n}}(\mathbf{s}, \omega_{\sigma 1}, \cdots, \omega_{\sigma n}) \\ &\qquad \otimes \mathbf{e}^{i_1} \otimes \cdots \otimes \mathbf{e}^{i_n}, \end{aligned} \tag{8}$$

and to deduce the following expression of the polarization vector of the n-th order:

$$\mathbf{P}^{(n)}(\mathbf{s}, t) = \varepsilon_0 \int_{-\infty}^{\infty} d\omega_1 \cdots \int_{-\infty}^{\infty} d\omega_n \, \chi^{(n)}(\mathbf{s}, \omega_1, \cdots, \omega_n) \tag{9}$$
$$\hat{\mathbf{E}}(\mathbf{s}, \omega_1) \cdots \hat{\mathbf{E}}(\mathbf{s}, \omega_n) e^{-i(\omega_1 + \cdots + \omega_n)t},$$

with a symmetric $\chi^{(n)}$ in the sense that

$$\chi^{(n)\,i}{}_{i_{\tau 1} \cdots\, i_{\tau n}}(\mathbf{s}, \omega_{\tau 1}, \cdots, \omega_{\tau n}) = \chi^{(n)\,i}{}_{i_1 \cdots\, i_n}(\mathbf{s}, \omega_1, \cdots, \omega_n), \quad \forall \tau \in \mathcal{S}_n.$$

This symmetry of the (not underlined) susceptibility tensor is called the intrinsic permutation symmetry. It allows for example to consider only sets of angular frequencies $(\omega_1, \cdots, \omega_n)$ ordered such that $\omega_1 \leqslant \omega_2 \leqslant \cdots \leqslant \omega_n$.

Finally, since the zero-th order susceptibility tensor does not present degeneracy, we define $\chi^{(0)} := \underline{\chi}^{(0)}$.

The Hermitian symmetry The electric field (and the polarization vector field) $\mathbf{E}$ being real, the harmonic components satisfy $\hat{\mathbf{E}}(\mathbf{s}, \omega) = \overline{\hat{\mathbf{E}}}(\mathbf{s}, -\omega)$ for all $\mathbf{s}$ in $\mathbb{R}^3$. This leads to the Hermitian symmetry of the susceptibility tensor fields:

$$\overline{\chi^{(n)}}(\mathbf{s}, \omega_1, \cdots, \omega_n) = \chi^{(n)}(\mathbf{s}, -\omega_1, \cdots, -\omega_n).$$

3 The propagation equation systems

3.1 The general propagation equation system

The magnetic response The preceding section shows how the electric answer of a medium is encoded in the (electric) susceptibility tensors. A similar treatment shows how the magnetic answer of a medium is encoded in the magnetic susceptibility tensors. Nevertheless, for the sake of simplicity of this paper, and also because at the wavelength we are looking at, nonlinear magnetic effects are usually negligible, we suppose from now on that the magnetic characteristics of the media are smooth, linear, local in space and stationary. This means that the magnetic inductive functional has the form

$$\mathbf{H}(\mathbf{s},t) = \mathfrak{H}\big(\{\mathbf{B}(\mathbf{s},\tau)\}_{\tau\in T_b^h(t)},\mathbf{s}\big)$$

so that

$$\mathbf{P}_m(\mathbf{s},t) = \mu_0^{-1}\int_{-\infty}^{\infty} d\omega\,\chi_m^{(1)}(\mathbf{s},\omega)\hat{\mathbf{B}}(\mathbf{s},\omega)e^{-i\omega t}.$$

To recover the usual conventions, we define the relative permittivity by:

$$\hat{\mu}_r(\mathbf{s},\omega) = (1+\chi_m^{(1)}(\mathbf{s},\omega))^{-1},$$

so that

$$\hat{\mathbf{B}} = \hat{\mu}\hat{\mathbf{H}}. \tag{10}$$

This last equation closes the sets of Maxwell's equations. Reporting the expressions of the inductive vector fields in terms of the primitive fields ((9) as well as the development of the polarization vector field in the definition 1 for the electric response, (10) for the magnetic response) in the first set of Maxwell's equations leads to the propagation equations. This section is devoted to present them, and to suggest some approximations in order to be able to implement them in a computer program.

The general propagation equation system On physical grounds, we suppose that the permeability tensor never vanishes so that the two vectorial Maxwell's equations lead to

$$\nabla\times\big(\hat{\mu}^{-1}(\mathbf{s},\omega)\nabla\times\hat{\mathbf{E}}(\mathbf{s},\omega)\big) - \omega^2\hat{\mathbf{D}}(\mathbf{s},\omega) = i\omega\hat{\mathbf{J}}(\mathbf{s},\omega) \tag{11}$$

for all ω in $\mathbb{R}$ and $\mathbf{s}$ in $\mathbb{R}^3$.

The dependence of $\mathbf{D}$ in $\mathbf{E}$ and in the (electric) characteristic of the medium were obtained in the first section, so that we just have to insert them in this expression. For this we have to write the polarization vector in the Fourier basis:

$$
\begin{aligned}
\hat{\mathbf{P}}^{(n)}(\mathbf{s},\omega) &= \frac{1}{2\pi}\int_{-\infty}^{\infty} dt\,\mathbf{P}^{(n)}(\mathbf{s},t)e^{i\omega t} \\
&= \frac{\varepsilon_0}{2\pi}\int_{-\infty}^{\infty} dt \int_{-\infty}^{\infty} d\omega_1 \cdots \int_{-\infty}^{\infty} d\omega_n\,\chi^{(n)}(\mathbf{s},\omega_1,\cdots,\omega_n) \\
&\quad \hat{\mathbf{E}}(\mathbf{s},\omega_1)\cdots\hat{\mathbf{E}}(\mathbf{s},\omega_n)e^{-i(\omega_1+\cdots+\omega_n-\omega)t}, \\
&= \varepsilon_0 \int_{-\infty}^{\infty} d\omega_1 \cdots \int_{-\infty}^{\infty} d\omega_n\,\chi^{(n)}(\mathbf{s},\omega_1,\cdots,\omega_n) \\
&\quad \hat{\mathbf{E}}(\mathbf{s},\omega_1)\cdots\hat{\mathbf{E}}(\mathbf{s},\omega_n)\delta(\omega_1+\cdots+\omega_n-\omega)
\end{aligned}
$$

where δ is the Dirac distribution; the last equation follows upon integration upon the time variable. For the spontaneous polarization vector, we have

$$
\hat{\mathbf{P}}^{(0)}(\mathbf{s},\omega) = \varepsilon_0\chi^{(0)}(\mathbf{s})\delta(\omega).
$$

The propagation equation system is therefore

$$
\begin{gathered}
\nabla \times \left(\hat{\mu}^{-1}(\mathbf{s},\omega)\nabla\times\hat{\mathbf{E}}(\mathbf{s},\omega)\right) - \omega^2\varepsilon_0\Big(\hat{\mathbf{E}}(\mathbf{s},\omega) + \chi^{(0)}(\mathbf{s})\delta(\omega) \\
+\sum_{n\in\mathbb{N}}\int_{-\infty}^{\infty} d\omega_1 \cdots \int_{-\infty}^{\infty} d\omega_n\,\chi^{(n)}(\mathbf{s},\omega_1,\cdots,\omega_n)\hat{\mathbf{E}}(\mathbf{s},\omega_1)\cdots\hat{\mathbf{E}}(\mathbf{s},\omega_n) \\
\delta(\omega_1+\cdots+\omega_n-\omega)\Big) = i\omega\hat{\mathbf{J}}(\mathbf{s},\omega).
\end{gathered} \tag{12}
$$

3.2 Nonlinearity of the second order

The introduction of some notations Of course, the propagation equation system (12) is too complicated to be solved directly. We will thus suppose from now on that no nonlinearity higher than the quadratic one are present. We will treat this order of nonlinearity with some detail; the other ones being direct but more and more sophisticated generalizations of this case. Also, though its presence does not lead to high difficulties, we will neglect the spontaneous polarization. The propagation equation system thus becomes

$$
\begin{gathered}
\nabla \times \left(\hat{\mu}^{-1}(\mathbf{s},\omega)\nabla\times\hat{\mathbf{E}}(\mathbf{s},\omega)\right) \\
-\omega^2\varepsilon_0\Big(\hat{\mathbf{E}}(\mathbf{s},\omega) + \int_{-\infty}^{\infty} d\omega_1\,\chi^{(1)}(\mathbf{s},\omega_1)\hat{\mathbf{E}}(\mathbf{s},\omega_1)\delta(\omega_1-\omega) \\
+\int_{-\infty}^{\infty} d\omega_1 \int_{-\infty}^{\infty} d\omega_2\,\chi^{(2)}(\mathbf{s},\omega_1,\omega_2)\hat{\mathbf{E}}(\mathbf{s},\omega_1)\hat{\mathbf{E}}(\mathbf{s},\omega_2)\delta(\omega_1+\omega_2-\omega)\Big) \\
= i\omega\hat{\mathbf{J}}(\mathbf{s},\omega)
\end{gathered}
$$

for all ω in $\mathbb{R}$ and $\mathbf{s}$ in $\mathbb{R}^3$.

In order to facilitate the reading of these equations, we introduce some notations. First, the usual linear permittivity: $\widehat{\varepsilon_r}^{(1)}(\mathbf{s},\omega) := 1+\chi^{(1)}(\mathbf{s},\omega)$. Using this and integrating the Dirac distributions, we have

$$\nabla\times\big(\hat{\mu}^{-1}(\mathbf{s},\omega)\nabla\times\hat{\mathbf{E}}(\mathbf{s},\omega)\big)-\omega^2\varepsilon_0\Big(\widehat{\varepsilon_r}^{(1)}(\mathbf{s},\omega)\hat{\mathbf{E}}(\mathbf{s},\omega)$$
$$+\int_{-\infty}^{\infty}d\omega_1\,\chi^{(2)}(\mathbf{s},\omega_1,\omega-\omega_1)\hat{\mathbf{E}}(\mathbf{s},\omega_1)\hat{\mathbf{E}}(\mathbf{s},\omega-\omega_1)\Big)=i\omega\hat{\mathbf{J}}(\mathbf{s},\omega)$$

for all ω in $\mathbb{R}$ and $\mathbf{s}$ in $\mathbb{R}^3$.

Then, we define the linear Maxwell's operator by

$$\mathcal{M}^{lin}_{(\mathbf{s},\omega)}\big(\hat{\mathbf{E}}(\mathbf{s},\omega)\big):=-\varepsilon_0^{-1}\nabla\times\big(\hat{\mu}^{-1}(\mathbf{s},\omega)\nabla\times\hat{\mathbf{E}}(\mathbf{s},\omega)\big)+\omega^2\widehat{\varepsilon_r}^{(1)}(\mathbf{s},\omega)\hat{\mathbf{E}}(\mathbf{s},\omega);$$

this operator is defined such that, in a linear media, we have

$$\mathcal{M}^{lin}_{(\mathbf{s},\omega)}\big(\hat{\mathbf{E}}(\mathbf{s},\omega)\big)=\frac{-i\omega}{\varepsilon_0}\hat{\mathbf{J}}(\mathbf{s},\omega)$$

for all ω in $\mathbb{R}$ and $\mathbf{s}$ in $\mathbb{R}^3$. Finally, we will use the notation

$$\lfloor\hat{\mathbf{E}}(\mathbf{s},\omega_1),\cdots,\hat{\mathbf{E}}(\mathbf{s},\omega_n)\rfloor:=\chi^{(n)}(\mathbf{s},\omega_1,\cdots,\omega_n)\hat{\mathbf{E}}(\mathbf{s},\omega_1)\cdots\hat{\mathbf{E}}(\mathbf{s},\omega_n);$$

this term is the contribution to the polarization vector $\hat{\mathbf{P}}(\mathbf{s},\omega_1+\cdots+\omega_n)$ of the interaction between the vectors $\hat{\mathbf{E}}(\mathbf{s},\omega_1)$, $\cdots$, $\hat{\mathbf{E}}(\mathbf{s},\omega_n)$. The propagation equation system is now

$$\mathcal{M}^{lin}_{(\mathbf{s},\omega)}\big(\hat{\mathbf{E}}(\mathbf{s},\omega)\big)+\omega^2\int_{-\infty}^{\infty}d\omega_1\,\lfloor\hat{\mathbf{E}}(\mathbf{s},\omega_1),\hat{\mathbf{E}}(\mathbf{s},\omega-\omega_1)\rfloor=\frac{-i\omega}{\varepsilon_0}\hat{\mathbf{J}}(\mathbf{s},\omega)\qquad(13)$$

for all ω in $\mathbb{R}$ and $\mathbf{s}$ in $\mathbb{R}^3$.

The harmonic assumption and the definition of the degree We focus on the case where the incident vector field is monochromatic: $\mathbf{E}^i(\mathbf{s},t)=\mathbf{E}^i_1(\mathbf{s})e^{-i\omega_I t}+\mathbf{E}^i_{-1}(\mathbf{s})e^{i\omega_I t}$ (we note $\mathbf{E}^i_p(\mathbf{s})$ for $\hat{\mathbf{E}}^i(\mathbf{s},p\omega_I)$). To simplify the system (13), which is described by a continuous parameter, we suppose that the susceptibility tensor field is nonzero only when it is evaluated on the harmonics of the incident angular frequency

$$\chi^{(n)}(\mathbf{s},\omega_1,\cdots,\omega_n)=0$$

if there is a j in $\{1,\cdots,n\}$ such that no integer p satisfies $\omega_j=p\omega_I$. This is an important hypothesis, which implies that we will concentrate on the harmonic generations and neglect the Raman or Brilloin scatterings, or subharmonic generations. On one hand, we have to admit that this is a serious stretch to the general aim stated in the introduction. On the other hand, the set of harmonic generations is sufficiently important and large to pursue this study.

The propagation equation of the p-th harmonic is now (we stop to explicitly write the point $\mathbf{s}$ where the fields are evaluated - note that no confusion can appear, because of the locality of the considered media; this is just a return to equalities between vector fields on $\mathbb{R}^3$ and not simply vectors in $\mathbb{R}^3$ - we also write $\mathcal{M}_p^{lin}$ for $\mathcal{M}_{(\mathbf{s},p\omega_I)}^{lin}$)

$$\mathcal{M}_p^{lin}(\mathbf{E}_p) + (p\omega_I)^2 \sum_{q\in\mathbb{Z}} \lfloor \mathbf{E}_q, \mathbf{E}_{p-q} \rfloor = \frac{-i\,p\omega_I}{\varepsilon_0} \mathbf{J}_p, \tag{14}$$

for all p in $\mathbb{Z}$. Since the sources oscillates only at the angular frequency ω_I (and of course $-\omega_I$), we have $\mathbf{J}_p = \mathbf{J}_p \delta_{|p|,1}$. We note that the harmonic assumption turns the propagation equation system from a system described by a parameter ranging on a *continuous* set to a system described by a parameter ranging on a *discrete* set.

Nevertheless, (14) is still too complicated to be solved: it still contains an *infinite* number of equations. To this aim, we introduce the notion of the **degree**, whose effect will be to obtain propagation equation systems described by a parameter ranging on a *finite* set. No absolute definition of the degree does exist; indeed, the way we will simplify the system (14) will depend on our purpose. To illustrate the choice we have to settle on, we give two definitions, and will argue that these two definitions present interesting, but different points of view.

Definition 2 *the degree d_1 of the approximation at the n-th order of a monochromatic field is*

$$d_1 := \min_{\underline{d}\in\mathbb{N}} \{ ||(p_1, \cdots, p_n)||_{l_1(\mathbb{Z})} > \underline{d} \Rightarrow \chi^{(n)}(\mathbf{s}; p_1\omega_I, \cdots, p_n\omega_I) = 0, \forall \mathbf{s} \in \mathbb{R}^3 \}$$

Definition 3 *the degree d_∞ of the approximation at the n-th order of a monochromatic field is*

$$\begin{aligned} d_\infty := \min_{\underline{d}\in\mathbb{N}} \{ & ||(p_1, \cdots, p_n, p_1 + \cdots + p_n)||_{l_\infty(\mathbb{Z})} > \underline{d} \\ & \Rightarrow \chi^{(n)}(\mathbf{s}; p_1\omega_I, \cdots, p_n\omega_I) = 0, \forall \mathbf{s} \in \mathbb{R}^3 \} \end{aligned}$$

With words, this means that, since we can consider only a finite number of components of the electric field, we suppose that the susceptibility tensors vanish if they are evaluated at high enough frequencies. In the d_1-th degree d, all the terms of the kind $\lfloor \mathbf{E}_{p_1}, \cdots, \mathbf{E}_{p_n} \rfloor$ vanish as soon as $|p_1| + \cdots + |p_n|$ is strictly greater than d. In the d_∞-th degree d, all the terms of the kind $\lfloor \mathbf{E}_{p_1}, \cdots, \mathbf{E}_{p_n} \rfloor$ vanish as soon as $|p_1|, \cdots,$ $|p_n|$ or $|p_1 + \cdots + p_n|$ is strictly greater than d. In both cases, the electric vector can be written, for all $\mathbf{s}$ in $\mathbb{R}^3$ and all t in $\mathbb{R}$ as

$$\begin{aligned} \mathbf{E}(\mathbf{s}, t) &= \sum_{\substack{p\in\mathbb{Z} \\ |p|\leqslant d}} \mathbf{E}_p(\mathbf{s}) e^{-ip\omega_I t} \\ &= \mathbf{E}_0 + 2\Re e\{ \sum_{1\leqslant p\leqslant d} \mathbf{E}_p(\mathbf{s}) e^{-ip\omega_I t} \}, \end{aligned}$$

since $\mathbf{E}_{-p} = \overline{\mathbf{E}_p}$.

We hope that these definitions of the degree will become intuitive when reading the next subsection.

3.3 The propagation equation systems in the lowest degree

We add a new hypothesis: the static component of the electric field $\mathbf{E}_0$ vanishes. Keeping it would not have brought huge difficulties, at least in this theoretical work. We note that this prevents us from studying the Pockels effect or the optical rectification.

3.3.1 In the degree 1 (linear case)

The cases $d_1 = 1$ and $d_\infty = 1$ are both:

$$\mathcal{M}_1^{lin}(\mathbf{E}_1) = \frac{-i\,\omega_I}{\varepsilon_0}\mathbf{J}_1.$$

This means that, whatever we use the d_1 or the d_∞ degree, the coarser approximation that we can do in nonlinear optics reduces to the linear case.

Remark 1 *The symmetry of the propagation equation system*

Due to the Hermitian symmetry of the Fourier components of the electric field ($\mathbf{E}_{-p} = \overline{\mathbf{E}_p}$), the one of the susceptibility tensor (see the paragraph The Hermitian symmetry*, page 9) and the one of the permeability tensor ($\mu(\mathbf{s}, -\omega) = \overline{\mu(\mathbf{s}, \omega)}$), it can be shown that the system (14) is coherent in the following sense: if $\mathbf{E}_p$ is a solution of the propagation equation for $\mathbf{E}_p$, then $\overline{\mathbf{E}_p}$ is a solution of the propagation equation for $\mathbf{E}_{-p}$. This allows to give only the equations for positive frequencies.*

3.3.2 In the degree 2 (second harmonic generation)

With the d_1 definition In the $d_1 = 2$ case, we have

$$\mathcal{M}_1^{lin}(\mathbf{E}_1) = \frac{-i\,\omega_I}{\varepsilon_0}\mathbf{J}_1, \tag{15a}$$

$$\mathcal{M}_2^{lin}(\mathbf{E}_2) + (2\omega_I)^2 \lfloor \mathbf{E}_1, \mathbf{E}_1 \rfloor = 0. \tag{15b}$$

Indeed, in this degree $d_1 = 2$, $\chi^{(2)}$ can be contracted only with the components $\mathbf{E}_p$ and $\mathbf{E}_q$ such that $|p| + |q| \leqslant 2$; since $\mathbf{E}_0 = 0$, we also have $p \neq 0$ and $q \neq 0$. Hence, the nonzero susceptibility tensors are $\chi^{(2)}(-\omega_I, -\omega_I)$, $\chi^{(2)}(\omega_I, -\omega_I)$, $\chi^{(2)}(-\omega_I, \omega_I)$ et $\chi^{(2)}(\omega_I, \omega_I)$. But the second and the third ones do not appear in the propagation equation system since we suppose that $\mathbf{E}_0$ vanishes, and the first one describes the generation of the component $\mathbf{E}_{-2}$, that we directly obtain from the equation that satisfies $\mathbf{E}_2$.

This propagation equation system has the advantage that it can be solved treating only linear equations: one first solves the equation that $\mathbf{E}_1$ satisfies and then the one

that $\mathbf{E}_2$ satisfies, treating $\mathbf{E}_1$ as a source. But this does not mean that the system is linear: if the incident field is multiplied by a constant factor m, $\mathbf{E}_1$ is scaled by m whereas $\mathbf{E}_2$ is scaled by m^2.

Since the equation that satisfies $\mathbf{E}_1$ is linear, it is commonly said that we are in the framework of the nondepletion of the pump beam. Indeed, $\mathbf{E}_1$ can be seen as a tank: the fact that it generates $\mathbf{E}_2$ does not change its energy.

With the d_∞ definition In the $d_\infty = 2$ case, we have

$$\mathcal{M}_1^{lin}(\mathbf{E}_1) + 2\omega_I^2 \lfloor \mathbf{E}_{-1}, \mathbf{E}_2 \rfloor = \frac{-i\,\omega_I}{\varepsilon_0}\mathbf{J}_1, \tag{16a}$$

$$\mathcal{M}_2^{lin}(\mathbf{E}_2) + (2\omega_I)^2 \lfloor \mathbf{E}_1, \mathbf{E}_1 \rfloor = 0. \tag{16b}$$

Considering this system, the definition of the degree d_∞ can appear to be nonphysical: in the generic case, that is without specifying a material, there are no reason for keeping the term $\lfloor \mathbf{E}_{-1}, \mathbf{E}_2 \rfloor$ when neglecting $\lfloor \mathbf{E}_1, \mathbf{E}_2 \rfloor$ that could generate the third harmonic (we recall that $\|\mathbf{E}_{-1}\| = \|\mathbf{E}_1\|$). This remark argues in the sense that the degree d_1 is the most natural one from the physical point of view. Nevertheless, in practical cases, the degree d_∞ can be important since it allows to consider a lossless medium - a fact, as seen above, impossible with the d_1 definition. The details are given in the section 4

3.3.3 In the degree 3 (third harmonic generation by a cascade effect)

A small number of interactions was considered in the degree 2. In the degree 3, we consider the interactions that result from a cascade effect. With the d_1-degree, the interactions between $\mathbf{E}_{-2}$ and $\mathbf{E}_2$ on one side, and the components $\mathbf{E}_{-1}$ and $\mathbf{E}_1$ on the other side are taken into account. With the d_∞-degree, we consider all the interactions where $\mathbf{E}_p$ occur for p in $\{-3, -2, -1, 1, 2, 3\}$.

With the d_1 definition In the $d_1 = 3$ case, we have

$$\mathcal{M}_1^{lin}(\mathbf{E}_1) + \omega_I^2 \Big(\lfloor \mathbf{E}_{-1}, \mathbf{E}_2 \rfloor + \lfloor \mathbf{E}_2, \mathbf{E}_{-1} \rfloor \Big) = \frac{-i\,\omega_I}{\varepsilon_0}\mathbf{J}_1,$$

$$\mathcal{M}_2^{lin}(\mathbf{E}_2) + (2\omega_I)^2 \lfloor \mathbf{E}_1, \mathbf{E}_1 \rfloor = 0,$$

$$\mathcal{M}_3^{lin}(\mathbf{E}_3) + (3\omega_I)^2 \Big(\lfloor \mathbf{E}_1, \mathbf{E}_2 \rfloor + \lfloor \mathbf{E}_2, \mathbf{E}_1 \rfloor \Big) = 0.$$

Using the intrinsic permutation symmetry, the propagation equation system gets

$$\mathcal{M}_1^{lin}(\mathbf{E}_1) + 2\omega_I^2 \lfloor \mathbf{E}_{-1}, \mathbf{E}_2 \rfloor = \frac{-i\,\omega_I}{\varepsilon_0}\mathbf{J}_1,$$

$$\mathcal{M}_2^{lin}(\mathbf{E}_2) + (2\omega_I)^2 \lfloor \mathbf{E}_1, \mathbf{E}_1 \rfloor = 0,$$

$$\mathcal{M}_3^{lin}(\mathbf{E}_3) + 2(3\omega_I)^2 \lfloor \mathbf{E}_1, \mathbf{E}_2 \rfloor = 0.$$

With the d_∞ definition In the $d_\infty = 3$ case, we have

$$\mathcal{M}_1^{lin}(\mathbf{E}_1) + 2\omega_I^2 \Big(\lfloor \mathbf{E}_{-2}, \mathbf{E}_3 \rfloor + \lfloor \mathbf{E}_{-1}, \mathbf{E}_2 \rfloor \Big) = \frac{-i\,\omega_I}{\varepsilon_0}\mathbf{J}_1,$$

$$\mathcal{M}_2^{lin}(\mathbf{E}_2) + (2\omega_I)^2 \Big(2\lfloor \mathbf{E}_{-1}, \mathbf{E}_3 \rfloor + \lfloor \mathbf{E}_1, \mathbf{E}_1 \rfloor \Big) = 0,$$

$$\mathcal{M}_3^{lin}(\mathbf{E}_3) + 2(3\omega_I)^2 \lfloor \mathbf{E}_1, \mathbf{E}_2 \rfloor = 0.$$

It may seem curious that we consider the third harmonic generation without taking into account the third order polarization. In fact, we advise to the reader not to use these systems - they were exposed only to give better intuition on the notion of degree.

In the second order of nonlinearity, the relevant systems are the ones obtain in the degree 2. It is a general fact that the n-th harmonic generation is to be studied in the n-th order of nonlinearity and in the n-th degree (at least with the two definition of the degree presented here).

3.4 Nonlinearity of the third order

The remark given in the second order of nonlinearity and in the third degree argues that we have to take into account the third order polarization vector field to treat the third harmonic generation. Roughly speaking, the study of the nonlinearity of the second order was aimed for presenting our method. The 'interesting' propagation equation system, (15) and (16), show little interaction. In the third order, more interactions are considered, so that the physical effects are richer: the third harmonic generation, the optical Kerr-effect, cascade effects. To study Raman scattering, one has to generalize the result of this subsection to non-harmonic processes, as in done in [9].

A treatment similar to the one used to obtain the system (14) has to be done with now $\mathbf{P} = \mathbf{P}^{(1)} + \mathbf{P}^{(2)} + \mathbf{P}^{(3)}$. The propagation equation system we obtain is

$$\mathcal{M}_p^{lin}(\mathbf{E}_p) + (p\omega_I)^2\Big(\sum_{q\in\mathbb{Z}}\lfloor\mathbf{E}_q, \mathbf{E}_{p-q}\rfloor + \sum_{(q,r)\in\mathbb{Z}^2}\lfloor\mathbf{E}_q, \mathbf{E}_r, \mathbf{E}_{p-q-r}\rfloor\Big) = \frac{-i\,p\omega_I}{\varepsilon_0}\mathbf{J}_p. \tag{19}$$

In agreement with the last paragraph of the last subsection, the interesting cases in the third order of nonlinearity are when the degree is also equal to three. We will thus consider only these cases. It is still assumed that the incident field is monochromatic, so that $\mathbf{J}_p = \mathbf{J}_p\delta_{p,|1|}$.

With the d_1 definition In the $d_1 = 3$ case, we have

$$\mathcal{M}_1^{lin}(\mathbf{E}_1) + \omega_I^2\Big(2\lfloor\mathbf{E}_{-1}, \mathbf{E}_2\rfloor + 3\lfloor\mathbf{E}_{-1}, \mathbf{E}_1, \mathbf{E}_1\rfloor\Big) = \frac{-i\,\omega_I}{\varepsilon_0}\mathbf{J}_1, \tag{20a}$$

$$\mathcal{M}_2^{lin}(\mathbf{E}_2) + (2\omega_I)^2\lfloor\mathbf{E}_1, \mathbf{E}_1\rfloor = 0, \tag{20b}$$

$$\mathcal{M}_3^{lin}(\mathbf{E}_3) + (3\omega_I)^2\Big(2\lfloor\mathbf{E}_1, \mathbf{E}_2\rfloor + \lfloor\mathbf{E}_1, \mathbf{E}_1, \mathbf{E}_1\rfloor\Big) = 0. \tag{20c}$$

This system presents several effects: the term $\lfloor\mathbf{E}_{-1}, \mathbf{E}_2\rfloor$ describes the counteraction of $\mathbf{E}_2$ on $\mathbf{E}_1$ (as it appears with the second order of nonlinearity and the degree $d_\infty = 2$), the pump beam is thus depleted; $\lfloor\mathbf{E}_{-1}, \mathbf{E}_1, \mathbf{E}_1\rfloor$ describes the optical Kerr-effect; lastly, the third harmonic is generated by two different processes: the cascade effect $\lfloor\mathbf{E}_1, \mathbf{E}_2\rfloor$, and the third-order process $\lfloor\mathbf{E}_1, \mathbf{E}_1, \mathbf{E}_1\rfloor$.

With the d_∞ definition In the $d_\infty = 3$ case, we have

$$\mathcal{M}_1^{lin}(\mathbf{E}_1) \tag{21a}$$
$$+ \omega_I^2 \Big(2\lfloor \mathbf{E}_{-2}, \mathbf{E}_3 \rfloor + 2\lfloor \mathbf{E}_{-1}, \mathbf{E}_2 \rfloor$$
$$+ 6\lfloor \mathbf{E}_{-3}, \mathbf{E}_1, \mathbf{E}_3 \rfloor + 3\lfloor \mathbf{E}_{-1}, \mathbf{E}_{-1}, \mathbf{E}_3 \rfloor + 3\lfloor \mathbf{E}_{-3}, \mathbf{E}_2, \mathbf{E}_2 \rfloor$$
$$+ 6\lfloor \mathbf{E}_{-2}, \mathbf{E}_1, \mathbf{E}_2 \rfloor + 3\lfloor \mathbf{E}_{-1}, \mathbf{E}_1, \mathbf{E}_1 \rfloor \Big) = \frac{-i\,\omega_I}{\varepsilon_0}\mathbf{J}_1,$$

$$\mathcal{M}_2^{lin}(\mathbf{E}_2) \tag{21b}$$
$$+ (2\omega_I)^2 \Big(2\lfloor \mathbf{E}_{-1}, \mathbf{E}_3 \rfloor + \lfloor \mathbf{E}_1, \mathbf{E}_1 \rfloor$$
$$+ 6\lfloor \mathbf{E}_{-3}, \mathbf{E}_2, \mathbf{E}_3 \rfloor + 6\lfloor \mathbf{E}_{-2}, \mathbf{E}_1, \mathbf{E}_3 \rfloor + 3\lfloor \mathbf{E}_{-2}, \mathbf{E}_2, \mathbf{E}_2 \rfloor$$
$$+ 6\lfloor \mathbf{E}_{-1}, \mathbf{E}_1, \mathbf{E}_1 \rfloor \Big) = 0,$$

$$\mathcal{M}_3^{lin}(\mathbf{E}_3) \tag{21c}$$
$$+ (3\omega_I)^2 \Big(2\lfloor \mathbf{E}_1, \mathbf{E}_2 \rfloor$$
$$+ 3\lfloor \mathbf{E}_{-3}, \mathbf{E}_3, \mathbf{E}_3 \rfloor + 6\lfloor \mathbf{E}_{-2}, \mathbf{E}_2, \mathbf{E}_3 \rfloor + 6\lfloor \mathbf{E}_{-1}, \mathbf{E}_1, \mathbf{E}_3 \rfloor$$
$$+ 3\lfloor \mathbf{E}_{-1}, \mathbf{E}_2, \mathbf{E}_2 \rfloor + \lfloor \mathbf{E}_1, \mathbf{E}_1, \mathbf{E}_1 \rfloor \Big) = 0.$$

The optical effects that this systems describes are the same than the ones present in the $d_1 = 3$ degree: optical Kerr-effect, depletion of the pump beam, second and third harmonic generations.

Once again, only energy considerations, presented in the following section, can justify that we keep terms like $\lfloor \mathbf{E}_2, \mathbf{E}_2, \mathbf{E}_{-3} \rfloor$, that are, of course, several orders of magnitude smaller than $\lfloor \mathbf{E}_1, \mathbf{E}_1, \mathbf{E}_{-1} \rfloor$ (see the equation (21a)).

This last example closes this section. The introduction of the degree allows to present systems whose solutions can be numerically investigated. Moreover, we see that, from the physical point of view (i.e., with the degree d_1), the optical Kerr effect appears simultaneously to the depletion of the pump beam or the third harmonic generation. This kind of statements is exactly the one we looked for; this is why we consider that, when restricting to harmonic processes with only one angular frequency in the incident field, we have fulfil our general aim. In [9] is exposed a method to generalize this when the scattered field oscillate at a nonharmonic frequencies or when the source oscillate at two frequencies. But this presents several drawbacks: first, this does not allow to treat phenomena described by a continuous set of frequencies, which are of course the most interesting ones from the physical point of view, and secondly, the systems obtained quickly become huge; solving them requires new hypotheses, so that it is like going back to the starting point: we have to impose constraints that depend on the particular situation we want to study.

Remark 2 *The optical Kerr effect*

We said that the n-th harmonic generation is better studied in the n-th order of nonlinearity and the n-th degree; nevertheless, the third order of nonlinearity in the degree $d_\infty = 1$ is also interesting. It consists of the single equation

$$\mathcal{M}_1^{lin}(\mathbf{E}_1) + 3\omega_I^2 \lfloor \mathbf{E}_{-1}, \mathbf{E}_1, \mathbf{E}_1 \rfloor = \frac{-i\,\omega_I}{\varepsilon_0} \mathbf{J}_1. \tag{22}$$

This is the optical Kerr effect. We repeat that we can argue that this system discards the term $\lfloor \mathbf{E}_1, \mathbf{E}_1, \mathbf{E}_1 \rfloor$ while keeping $\lfloor \mathbf{E}_{-1}, \mathbf{E}_1, \mathbf{E}_1 \rfloor$, thus 'neglecting' a priori *that $\|\mathbf{E}_{-1}\| = \|\mathbf{E}_1\|$. Nevertheless, in some cases, we can be concerned only to what happens in the fundamental frequency, and so we disregard the harmonic generation. This naturally leads to the study of this optical Kerr effect. We note that, dealing with this equation, we neglect the counter-reaction of the harmonics on the fundamental component, i.e., we neglect the term:*

$$\begin{gathered} 2\lfloor \mathbf{E}_{-2}, \mathbf{E}_3 \rfloor + 2\lfloor \mathbf{E}_{-1}, \mathbf{E}_2 \rfloor + 6\lfloor \mathbf{E}_{-3}, \mathbf{E}_1, \mathbf{E}_3 \rfloor \\ +3\lfloor \mathbf{E}_{-1}, \mathbf{E}_{-1}, \mathbf{E}_3 \rfloor + 3\lfloor \mathbf{E}_{-3}, \mathbf{E}_2, \mathbf{E}_2 \rfloor + 6\lfloor \mathbf{E}_{-2}, \mathbf{E}_1, \mathbf{E}_2 \rfloor \end{gathered}$$

that contributes to $\hat{\mathbf{P}}(\omega_I)$. For non-centrosymmetric media, in which the second order polarization vector fields vanish, this approximation can be dangerous, since it is not always clear that

$$2\|\lfloor \mathbf{E}_{-1}, \mathbf{E}_2 \rfloor\| \ll 3\|\lfloor \mathbf{E}_{-1}, \mathbf{E}_1, \mathbf{E}_1 \rfloor\|.$$

However, for centrosymmetric media, the term $\|\lfloor \mathbf{E}_{-1}, \mathbf{E}_1, \mathbf{E}_1 \rfloor\|$ must be the leading one among the terms that contributes to $\hat{\mathbf{P}}^{(3)}(\omega_I)$; this justifies that the $d_\infty = 1$ system is an approximation of the $d_\infty = 3$ system. Hence, the way we obtained this equation shows that the optical Kerr effect appears naturally, and that it does make sense to study a harmonic *nonlinear effect.*

We will go back, in the next section, to that simple system as a basic example of energy conservation.

4 Electric energy considerations in nonlinear media

4.1 The stochastic mean of the electric power density

Before dealing with energy conservation in nonlinear optics, we have to give a rigorous definition of what we mean by energy transfer. Indeed, the components oscillating at the frequency of the incident field generate the other components, and so will loose some energy. The question is thus the following: when this harmonic generation occur with no loss of electric energy, that is, without Joule effect. Stated in an other way: when does the energy in the electric sources equal the energy in the scattered field?

To this aim, we go back to the definition of the energy-momentum quadrivector, or, more precisely, its density, denoted by $(c\mathcal{I}, W)$. By definition, it is divergence-free, so that in a Cartesian coordinate frame, where

$$\nabla^4 \cdot := (-c\partial_x \ \ -c\partial_y \ \ -c\partial_z \ \ \partial_t)\cdot,$$

we have

$$-c\,\nabla\cdot(c\,\mathcal{I})+\partial_t W=0.$$

From now on, we consider only the electromagnetic part $(c\,\mathcal{I}_{em},W_{em})$ of $(c\,\mathcal{I},W)$. We thus say that no transfer between electromagnetic energy-momentum and other form of energy-momentum occurs if

$$-c\,\nabla\cdot(c\,\mathcal{I}_{em})+\partial_t W_{em}=0.$$

For geometric reasons, and for the respect of the units of each field, the Poynting vector field, $\mathcal{P}:=\mathbf{E}\times\mathbf{H}$, is defined to be the density of electromagnetic momentum $c\,\mathcal{I}_{em}$ multiplied by the factor $-c$. The electromagnetic energy-momentum conservation is thus

$$\nabla\cdot\mathcal{P}+\partial_t W_{em}=0.$$

This relation allows to define the electromagnetic energy density W_{em}. Indeed, Maxwell's equations give:

$$\begin{aligned}\partial_t W_{em}&=-\nabla\cdot\mathcal{P}\\&=(\partial_t\mathbf{B})\cdot\mathbf{H}+(\partial_t\mathbf{D})\cdot\mathbf{E}+\mathbf{E}\cdot\mathbf{J}.\end{aligned}$$

We define the electric energy density W_e by

$$\partial_t W_e=(\partial_t\mathbf{D})\cdot\mathbf{E}.$$

We write the electric vector field evaluated at the point $\mathbf{s}$ and the time t as (we need an absolute convergence of this series: indeed, in this case, the series converge uniformly with respect to the time variable and thus we will be allowed to change the integral operation with the sum operation)

$$\mathbf{E}(\mathbf{s},t)=\sum_{p\in\mathbb{Z}}\mathbf{E}_p(\mathbf{s})e^{-ip\omega_I t}.$$

Once again, we will give all the details in the second order of nonlinearity, and then present more general formulae. So let us write

$$\begin{aligned}\mathbf{D}(t)&=\varepsilon_0\mathbf{E}(t)+\mathbf{P}(t)\\&=\varepsilon_0\sum_{p\in\mathbb{Z}}\Big(\mathbf{E}_p+\lfloor\mathbf{E}_p\rfloor+\sum_{q\in\mathbb{Z}}\lfloor\mathbf{E}_q,\mathbf{E}_{p-q}\rfloor\Big)e^{-ip\omega_I t}.\end{aligned}$$

The electric energy density is therefore[1]

[1] Up to now, the dot product was on $\mathbb{R}^3$. We extend it on $\mathbb{C}^3$ since we decompose the vectors in their Fourier components. We choose to take it linear in both variables, and so it will be symmetric - this will not bring problems since the nondegeneracy will never be exploited.

$$
\begin{aligned}
W_e(t) = W_e(0) & \\
&+ \varepsilon_0 \sum_{(p,q)\in\mathbb{Z}^*\times\mathbb{Z}} \frac{q}{p}\mathbf{E}_{p-q}\cdot\big(\mathbf{E}_q + \lfloor \mathbf{E}_q\rfloor + \sum_{r\in\mathbb{Z}}\lfloor \mathbf{E}_r, \mathbf{E}_{q-r}\rfloor\big)(e^{-ip\omega_I t}-1)\\
&+ \varepsilon_0 \sum_{p\in\mathbb{Z}} -ip\omega_I \mathbf{E}_{-p}\cdot\big(\mathbf{E}_p + \lfloor \mathbf{E}_p\rfloor + \sum_{q\in\mathbb{Z}}\lfloor \mathbf{E}_q, \mathbf{E}_{p-q}\rfloor\big)t.
\end{aligned}
$$

The first term in this expression is a constant without any physical significance, the second one is oscillating and the last one diverges linearly with t. For large t (large with respect to the period associated to the fundamental angular frequency ω_I), only the last term is relevant. This leads to define the following quantity, which is the stochastic mean of the electric power density:

$$
\begin{aligned}
< \partial_t W_e >: &= \lim_{T\to\infty}\frac{1}{T}\int_0^T dt\, \partial_t W_e\\
&= \varepsilon_0 \sum_{p\in\mathbb{Z}} \Big(-ip\omega_I \mathbf{E}_{-p}\cdot\big(\mathbf{E}_p + \lfloor \mathbf{E}_p\rfloor + \sum_{q\in\mathbb{Z}}\lfloor \mathbf{E}_q, \mathbf{E}_{p-q}\rfloor\big)\Big).
\end{aligned}
$$

The term $\varepsilon_0\big(\mathbf{E}_p + \lfloor \mathbf{E}_p\rfloor + \sum_{q\in\mathbb{Z}}\lfloor \mathbf{E}_q, \mathbf{E}_{p-q}\rfloor\big)$, is the expansion of $\mathbf{D}_p$ as $\varepsilon_0\mathbf{E}_p + \mathbf{P}_p^{(1)} + \mathbf{P}_p^{(2)}$. We recall that, by construction, the answer of the medium is described by the polarization vector field $\mathbf{P}$; this is coherent with the fact that the sum $\sum_{p\in\mathbb{Z}} -ip\omega_I\mathbf{E}_{-p}\cdot\mathbf{E}_p$ (where no susceptibility tensor appears) vanishes - we map $p\mapsto -p$ and use the Hermitian symmetry of the duality product on $\mathbb{C}^3$. We thus have shown that, in vacuum, the stochastic mean of the electric power density $\varepsilon_0\sum_{p\in\mathbb{Z}} -ip\omega_I\mathbf{E}_{-p}\cdot\mathbf{E}_p$ vanishes.

Going back to an arbitrary order of nonlinearity, we have

$$
\begin{aligned}
< \partial_t W_e > &= < (\partial_t \mathbf{P})\cdot\mathbf{E} > \qquad (23)\\
&= \varepsilon_0 \sum_{p\in\mathbb{Z}} \Big(-ip\omega_I \mathbf{E}_{-p}\cdot\big(\lfloor \mathbf{E}_p\rfloor + \sum_{q\in\mathbb{Z}}\lfloor \mathbf{E}_q, \mathbf{E}_{p-q}\rfloor + \cdots\big)\Big)\\
&= \sum_{n\in\mathbb{N}}\sum_{p\in\mathbb{Z}} -ip\omega_I\mathbf{E}_{-p}\cdot\mathbf{P}_p^{(n)}\\
&= \sum_{p\in\mathbb{Z}} -ip\omega_I\mathbf{E}_{-p}\cdot\mathbf{P}_p.
\end{aligned}
$$

We remark that this expression is a straightforward generalization of the stochastic mean of the electric power density in the linear regime. It is also clear that this expression is real - taking the complex conjugate is equivalent to map p to $-p$ (and q to $-q$ in the second line), and thus does not change the sum.

4.2 A symmetry of the susceptibility tensors and lossless media

We did not found a relation between the susceptibility tensors that exactly translate the property of a medium to be lossless. Nevertheless, we found one implication: if a certain symmetry between the susceptibility tensors is satisfied then the medium is lossless. This subsection is devoted to the presentation of this criterion.

The first hypothesis that we will do is to consider that the electromagnetic power density vanishes in each order (we won't repeat 'the stochastic mean of' each time, but it is always understood). We thus define

$$< \partial_t W_e^{(n)} >:= \sum_{p\in\mathbb{Z}} -ip\omega_I \mathbf{E}_{-p} \cdot \mathbf{P}_p^{(n)}.$$

The third equality in (23), shows that

$$< \partial_t W_e >= \sum_{n\in\mathbb{N}} < \partial_t W_e^{(n)} > .$$

Using the Hermitian symmetry of the Fourier component of the electric vector field and of the polarization vector field, it is straightforward to obtain the expression

$$< \partial_t W_e^{(n)} >= 2\omega_I \Im m\{\sum_{p\in\mathbb{N}} p\mathbf{E}_{-p} \cdot \mathbf{P}_p^{(n)}\}. \tag{24}$$

4.2.1 In the first order

In the first order, we have

$$\begin{aligned} < \partial_t W_e^{(1)} >: &= \varepsilon_0 \sum_{p\in\mathbb{Z}} -ip\omega_I \mathbf{E}_{-p} \cdot \lfloor \mathbf{E}_p \rfloor \\ &= -i\varepsilon_0\omega_I \sum_{p\in\mathbb{N}} p\mathbf{E}_{-p} \cdot \Big(\chi^{(1)}(p\omega_I) - \chi^{(1)\,T}(-p\omega_I)\Big)\mathbf{E}_p. \end{aligned}$$

Here we make the second assumption: we assume that each term of this sum vanishes. This leads to the conclusion that no electromagnetical energy is lost in the first order if the susceptibility tensor field (on $\mathbb{R}^3 \times \mathbb{R}$) $\chi^{(1)}$ is Hermitian, i.e.:

$$\chi^{(1)}(p\omega_I) = \chi^{(1)\,T}(-p\omega_I). \tag{25}$$

The conclusion is not exactly the well-known fact from undergraduate lessons: if the medium is linear - so that the fields oscillate at only one angular frequency ω_I -, it is lossless from the electrical point of view if and only if the susceptibility tensor field (on $\mathbb{R}^3$) $\chi^{(1)}(\cdot, \omega_I)$ is Hermitian. Here, the medium is nonlinear - so that the fields oscillate at every angular frequency $p\omega_I$ for p in $\mathbb{Z}^*$ -, and we showed that is lossless from the electrical point of view in the first order if the susceptibility tensor field $\chi^{(1)}$ is Hermitian.

4.2.2 In the second order

The general criterion We now go to the second order:

$$
\begin{aligned}
< \partial_t W_e^{(2)} > &= \sum_{p\in\mathbb{Z}} -ip\omega_I \mathbf{E}_{-p}\cdot \mathbf{P}_p^{(2)} \\
&= \varepsilon_0 \sum_{(p,q)\in\mathbb{Z}^2} -ip\omega_I \mathbf{E}_{-p}\cdot \lfloor \mathbf{E}_q, \mathbf{E}_{p-q}\rfloor .
\end{aligned}
$$

Some lines of computation show that

$$
< \partial_t W_e^{(2)} >= \omega_I \varepsilon_0 \Im m\{ \sum_{\substack{q\in\mathbb{N}\\ 0\leqslant r\leqslant q}} d_2(q,r) d_2(r,0) \mathbf{E}_{-(q+r)}\cdot \xi_{q,r}^{(2)} \mathbf{E}_r \mathbf{E}_q \},
$$

with

$$
\begin{aligned}
\xi_{q,r}^{(2)} := {} & r\big(\chi^{(2)}(r\omega_I, q\omega_I) - \chi^{(2)\,T_{12}}(-(q+r)\omega_I, q\omega_I)\big) \\
& + q\big(\chi^{(2)}(r\omega_I, q\omega_I) - \chi^{(2)\,T_{13}}(r\omega_I, -(q+r)\omega_I)\big).
\end{aligned}
$$

where the partial transposition notation is defined by $\mathbf{v}_a\cdot\chi^{(2)\,T_{12}}\mathbf{v}_b\mathbf{v}_c := \mathbf{v}_b\cdot\chi^{(2)}\mathbf{v}_a\mathbf{v}_c$ for all triples $(\mathbf{v}_a,\mathbf{v}_b,\mathbf{v}_c)$ of vectors in $\mathbb{R}^3$; with indices, this gives $(\chi^{(2)\,T_{12}})^i{}_{i_1 i_2} = \chi^{(2)}{}_{i_1}{}^{i}{}_{i_2}$. In the same way we write $\mathbf{v}_a\cdot\chi^{(2)\,T_{13}}\mathbf{v}_b\mathbf{v}_c := \mathbf{v}_c\cdot\chi^{(2)}\mathbf{v}_b\mathbf{v}_a$ and so $(\chi^{(2)\,T_{13}})^i{}_{i_1 i_2} = \chi^{(2)}{}_{i_2 i_1}{}^{i}$. Also, $d_2(\cdot,\cdot)$ is the degeneracy function of a set of two elements:

$$
d_2(a,b) = \begin{cases} 2, & a\neq b \\ 1, & a = b. \end{cases}
$$

For $< \partial_t W_e^{(2)} >$ to vanish, it is thus sufficient that each term of the sum vanishes (this is similar to what we did in the first order). We now introduce the third assumption: the factor of r and the one of q vanish independently. Thus, no transfer of electric energy to an other form of energy is possible if the tensor field $\chi^{(2)}$ satisfies, for any (q,r) in $\mathbb{Z}^2$ such that $q>0$ and $0\leqslant r\leqslant q$, the relations:

$$
\begin{cases}
\chi^{(2)}(r\omega_I, q\omega_I) = \chi^{(2)\,T_{12}}(-(q+r)\omega_I, q\omega_I) \\
\chi^{(2)}(r\omega_I, q\omega_I) = \chi^{(2)\,T_{13}}(r\omega_I, -(q+r)\omega_I).
\end{cases}
$$

A more symmetric way of writing these conditions appear if we define $p := -(q+r)$ and we write explicitly, as an argument of the susceptibility tensor, the angular frequency for which $\chi^{(2)}$ contribute in $\mathbf{P}^{(2)}$. Thus, from now on, any of the following expression has the same meaning: $\chi^{(n)}(\mathbf{s};\omega_0;\omega_1,\cdots,\omega_n)$, $\chi^{(n)}(\mathbf{s},\omega_1,\cdots,\omega_n)$, $\chi^{(n)}(\omega_0;\omega_1,\cdots,\omega_n)$, $\chi^{(n)}(\omega_1,\cdots,\omega_n)$, and when $\chi^{(n)}$ is evaluated on $n+1$ angular frequencies $\omega_0,\cdots,\omega_n$, it is understood that $\omega_0 = \omega_1+\cdots+\omega_n$. With this notation, a medium is lossless from the electrical point of view in the second order if

$$\chi^{(2)}(-p\omega_I;r\omega_I,q\omega_I)=\chi^{(2)\;T_{12}}(-r\omega_I;p\omega_I,q\omega_I)=\chi^{(2)\;T_{13}}(-q\omega_I;r\omega_I,p\omega_I). \tag{26}$$

So no electric energy loss is guaranteed if the symmetry that consists in permuting the indices together with the frequency variables holds for the $\chi^{(n)}$. This result appears in the book [10], but these authors do not consider the cascade effects that appear in harmonic generations. Moreover, the assumptions needed to derive these results are not explicitly given.

We note that (contrary to what is affirmed in [2], p.34) we can have a lossless medium with complex-valued susceptibility tensor fields. This has been checked numerically by the authors, with simulations similar to the ones given in [3].

Kleinman's relations If the medium is instantaneous, then the response function $R^{(n)}$ is of the kind $A \bigotimes_{i\in\{1,\cdots,n\}} \delta(t_i)$ where A is a function that depends only on the space variables. Then the susceptibility tensor fields do not depend on the angular frequencies:

$$\chi^{(n)}_{inst} := \chi^{(n)}(p_1\omega_I,\cdots,p_n\omega_I) \qquad \forall (p_1,\cdots,p_n)\in\mathbb{Z}^n.$$

Thus, in the first order, the polarization is

$$\begin{aligned}\mathbf{P}^{(1)}(t) &= \varepsilon_0\int_{-\infty}^{\infty} d\omega_1 \chi^{(1)}_{inst}\hat{\mathbf{E}}(\omega_1)e^{-i\omega_1 t}\\ &= \varepsilon_0\chi^{(1)}_{inst}\int_{-\infty}^{\infty} d\omega_1 \hat{\mathbf{E}}(\omega_1)e^{-i\omega_1 t}\\ &= \varepsilon_0\chi^{(1)}_{inst}\mathbf{E}(t).\end{aligned}$$

Since $\chi^{(1)}_{inst}$, when contracted with a real vector, gives a real vector, it is real.

In the same way, in the second order, we have

$$\begin{aligned}\mathbf{P}^{(2)}(t) &= \varepsilon_0\int_{-\infty}^{\infty} d\omega_1\int_{-\infty}^{\infty} d\omega_2\,\chi^{(2)}_{inst}\hat{\mathbf{E}}(\omega_1)\hat{\mathbf{E}}(\omega_2)e^{-i(\omega_1+\omega_2)t}\\ &= \varepsilon_0\chi^{(2)}_{inst}\int_{-\infty}^{\infty} d\omega_1\,\hat{\mathbf{E}}(\omega_1)e^{-i\omega_1 t}\int_{-\infty}^{\infty} d\omega_2\,\hat{\mathbf{E}}(\omega_2)e^{-i\omega_2 t}\\ &= \varepsilon_0\chi^{(2)}_{inst}\mathbf{E}(t)\mathbf{E}(t)\end{aligned}$$

and $\chi^{(2)}_{inst}$ is also real.

Reporting the criteria (25) and (26) in this context, we deduce that an instantaneous nonlinear medium is lossless if the susceptibility tensor fields that characterize it satisfy

$$\begin{cases}\text{in the first order :} & \chi^{(1)}_{inst}=\chi^{(1)\;T}_{inst}\\ \text{in the second order :} & \chi^{(2)}_{inst}=\chi^{(2)\;T_{12}}_{inst}=\chi^{(2)\;T_{13}}_{inst}.\end{cases}$$

Because the transposition (12) generate the symmetric group $\mathcal{S}_2$, the transpositions (12) and (13) generate $\mathcal{S}_3$, etc., where $\mathcal{S}_{n+1}$ acts as permutation of the components of $\chi^{(n)}$, we conclude that completely symmetric and frequency independent susceptibility tensor characterize lossless media. This is the famous result of D.A.Kleinman ([11]).

In the lower degrees In the second order $1 \leqslant n \leqslant 2$ and the degree $d_\infty = 2$, we have

$$\begin{aligned}
\mathbf{P}_1^{(1)} &= \varepsilon_0 \lfloor \mathbf{E}_1 \rfloor, \\
\mathbf{P}_2^{(1)} &= \varepsilon_0 \lfloor \mathbf{E}_2 \rfloor, \\
\mathbf{P}_1^{(2)} &= 2\varepsilon_0 \lfloor \mathbf{E}_{-1}, \mathbf{E}_2 \rfloor, \\
\mathbf{P}_2^{(2)} &= \varepsilon_0 \lfloor \mathbf{E}_1, \mathbf{E}_1 \rfloor.
\end{aligned}$$

Hence, an explicit expansion of (24) gives

$$< \partial_t W_e >= 2\varepsilon_0 \omega_I \Im m\{\mathbf{E}_{-1} \cdot (\lfloor \mathbf{E}_1 \rfloor + 2\lfloor \mathbf{E}_{-1}, \mathbf{E}_2 \rfloor) + 2\mathbf{E}_{-2} \cdot (\lfloor \mathbf{E}_2 \rfloor + \lfloor \mathbf{E}_1, \mathbf{E}_1 \rfloor)\}.$$

We first treat the first order of nonlinearity. So let us assume that $\chi^{(1)}(\mathbf{s}, \omega_I)$ is Hermitian for all $\mathbf{s}$ in $\mathbb{R}^3$, in the sense that its transpose conjugate is equal to itself; we then have

$$\begin{aligned}
\mathbf{E}_{-1} \cdot \lfloor \mathbf{E}_1 \rfloor :&= \mathbf{E}_{-1} \cdot \chi^{(1)}(\omega_I) \mathbf{E}_1 \\
&= \mathbf{E}_1 \cdot \chi^{(1)\,T}(\omega_I) \mathbf{E}_{-1} \\
&= \overline{\mathbf{E}_{-1}} \cdot \overline{\chi^{(1)}(\omega_I)}\,\overline{\mathbf{E}_1} \\
&= \overline{\mathbf{E}_{-1} \cdot \chi^{(1)}(\omega_I) \mathbf{E}_1} \\
&=: \overline{\mathbf{E}_{-1} \cdot \lfloor \mathbf{E}_1 \rfloor},
\end{aligned}$$

We deduce from this that $\Im m\{\mathbf{E}_{-1} \cdot \lfloor \mathbf{E}_1 \rfloor\}$ vanishes: this is the condition (25) on electrical energy conservation in the first order. A similar computation shows that if $\chi^{(1)}(\mathbf{s}, 2\omega_I)$ is Hermitian for all $\mathbf{s}$ in $\mathbb{R}^3$, then $\Im m\{\mathbf{E}_{-2} \cdot \lfloor \mathbf{E}_2 \rfloor\}$ vanishes.

Considering now the second order of nonlinearity, we suppose that the second order susceptibility tensor field satisfies

$$\chi^{(2)}(-p\omega_I; r\omega_I, q\omega_I) = \chi^{(2)\,T_{13}}(-q\omega_I; r\omega_I, p\omega_I) \tag{27}$$

for all (q, r) in $\mathbb{Z}^2$ - we will use only $\chi^{(2)\,T_{13}}(\omega_I; -\omega_I, 2\omega_I) = \chi^{(2)}(-2\omega_I; -\omega_I, -\omega_I)$. Then we have

$$
\begin{aligned}
< \partial_t W_e^{(2)} > &= 4\varepsilon_0\omega_I \Im m\{\mathbf{E}_{-1}\cdot\lfloor\mathbf{E}_{-1},\mathbf{E}_2\rfloor + \mathbf{E}_{-2}\cdot\lfloor\mathbf{E}_1,\mathbf{E}_1\rfloor\}\\
&= 4\varepsilon_0\omega_I \Im m\{\mathbf{E}_{-1}\cdot\chi^{(2)}(-\omega_I,2\omega_I)\mathbf{E}_{-1}\mathbf{E}_2 + \mathbf{E}_{-2}\cdot\chi^{(2)}(\omega_I,\omega_I)\mathbf{E}_1\mathbf{E}_1\}\\
&= 4\varepsilon_0\omega_I \Im m\{\mathbf{E}_2\cdot\chi^{(2)\ T_{13}}(-\omega_I,2\omega_I)\mathbf{E}_{-1}\mathbf{E}_{-1} + \mathbf{E}_{-2}\cdot\chi^{(2)}(\omega_I,\omega_I)\mathbf{E}_1\mathbf{E}_1\}\\
&= 4\varepsilon_0\omega_I \Im m\{\mathbf{E}_2\cdot\chi^{(2)}(-\omega_I,-\omega_I)\mathbf{E}_{-1}\mathbf{E}_{-1} + \mathbf{E}_{-2}\cdot\chi^{(2)}(\omega_I,\omega_I)\mathbf{E}_1\mathbf{E}_1\}\\
&= 8\varepsilon_0\omega_I \Im m\{\Re e\{\mathbf{E}_2\cdot\chi^{(2)}(-\omega_I,-\omega_I)\mathbf{E}_{-1}\mathbf{E}_{-1}\}\}\\
&= 0,
\end{aligned}
$$

where the last but one line is obtained by the Hermitian symmetry of the electric vector field and of the susceptibility tensor fields. We thus see that the criterion (27) is a sufficient condition for the stochastic mean of the electrical energy density at the second order to vanish.

This is why we argued, in the subsection 3.2, that though the degree d_1 is the most satisfying one from the physical point of view, even if it leads to consider $\mathbf{E}_1$ as a tank, the degree d_∞ allows to consider lossless media. This advantage can be a good test to check the coherence of numerical simulations.

Lastly, we note that (27) is weaker than (26); this is due to the degeneracy in the angular frequencies in the order two and degree two. The reader can check that the full criterion (26) is necessary if we consider the order two in the degree three.

4.2.3 In the third order

The general criterion In the third order, we have

$$
< \partial_t W_e^{(3)} > := \varepsilon_0 \sum_{(q,r,s)\in\mathbb{Z}^3} -iq\omega_I \mathbf{E}_{-q}\cdot\lfloor\mathbf{E}_r,\mathbf{E}_s,\mathbf{E}_{q-r-s}\rfloor.
$$

We can show ([9]) that if the following conditions are satisfied

$$
\begin{cases}
\chi^{(3)}(-p\omega_I;q\omega_I,r\omega_I,s\omega_I) = \chi^{(3)\ T_{12}}(-q\omega_I;p\omega_I,r\omega_I,s\omega_I)\\
\chi^{(3)}(-p\omega_I;q\omega_I,r\omega_I,s\omega_I) = \chi^{(3)\ T_{13}}(-r\omega_I;q\omega_I,p\omega_I,s\omega_I)\\
\chi^{(3)}(-p\omega_I;q\omega_I,r\omega_I,s\omega_I) = \chi^{(3)\ T_{14}}(-s\omega_I;q\omega_I,r\omega_I,p\omega_I)
\end{cases}
$$

then a medium is lossless from the electrical point of view in the third order.

In the lower degrees As exposed in the remark 2, the system of propagation equations (19) in the degree $d_\infty = 1$ reduces to the single equation that describes the optical Kerr effect:

$$
\mathcal{M}_1^{lin}(\mathbf{E}_1) + 3\omega_I^2\lfloor\mathbf{E}_{-1},\mathbf{E}_1,\mathbf{E}_1\rfloor = \frac{-i\,\omega_I}{\varepsilon_0}\mathbf{J}_1. \qquad ((22))
$$

Since

$$
\mathbf{P}_1^{(3)} = \varepsilon_0\lfloor\mathbf{E}_{-1},\mathbf{E}_1,\mathbf{E}_1\rfloor,
$$

the formula (24) gives

$$
\begin{aligned}
< \partial_t W_e^{(3)} >_{\text{Kerr}} &= 2\omega_I \Im m\{\sum_{p\in\mathbb{N}} p\mathbf{E}_{-p}\cdot \mathbf{P}_p^{(3)}\} \\
&= 6\varepsilon_0\omega_I \Im m\{\mathbf{E}_{-1}\cdot \lfloor \mathbf{E}_{-1}, \mathbf{E}_1, \mathbf{E}_1 \rfloor\}
\end{aligned}
$$

Assume furthermore that the medium is invariant along one axis (say (O,z)), and that if a beam linearly polarized along (O,z) impinges on this medium, then the scattered light is also polarized along (O,z). Then, the only components of the susceptibility tensors that matter are $\chi^{(1)\,z}{}_{z}$ and $\chi^{(3)\,z}{}_{zzz}$. Let us denote $\mathbf{E}_1(x,y,z)$ by $u_1(x,y)\hat{z}$.

In this case, we have

$$
\begin{aligned}
< \partial_t W_e^{(3)} >_{\text{Kerr}} &= 6\varepsilon_0\omega_I \Im m\{u_{-1}\chi^{(3)\,z}{}_{zzz}(-\omega_I,\omega_I,\omega_I)u_{-1}u_1u_1\} \\
&= 6\varepsilon_0\omega_I \Im m\{\chi^{(3)\,z}{}_{zzz}(-\omega_I,\omega_I,\omega_I)\}|u_1|^4 .
\end{aligned}
$$

In other words, with all these assumption, there are no electrical energy loss in the third order *if and only if* the susceptibility tensor $\chi^{(3)}$ is real-valued.

4.3 In the n-th order

The general proof that a sufficient condition for a medium to be lossless in the n-th order can be found in [9]. The result is that

$$
\begin{aligned}
< \partial_t W_e^{(n)} > &= \sum_{p\in\mathbb{Z}} -ip\omega_I \mathbf{E}_{-p}\cdot\mathbf{P}_p^{(n)} \\
&= -i\varepsilon_0\omega_I \sum_{(p_1,\cdots,p_n)\in\mathbb{Z}^n} (p_1+\cdots+p_n)\mathbf{E}_{-(p_1+\cdots+p_n)}\cdot\lfloor \mathbf{E}_{p_1},\cdots,\mathbf{E}_{p_n}\rfloor .
\end{aligned}
$$

vanishes if

$$
\begin{aligned}
\chi^{(n)\;T_{1\,j+1}}(-p_j\omega_I; p_0\omega_I,\cdots,p_{j-1}\omega_I,p_{j+1}\omega_I,\cdots,p_n\omega_I) = \\
\chi^{(n)}(-p_0\omega_I;p_1\omega_I,\cdots,p_n\omega_I) \qquad \forall j\in\{1,\cdots,n\}
\end{aligned}
$$

where $p_0 := -(p_1+\cdots+p_n)$. We can check that the criteria given for the lowest order fulfils this set of conditions.

Once again, we repeat that we did not find a necessary and sufficient criterion: we supposed that each order vanishes independently, that the term $< \partial_t(W_e^{(n)} >$ vanishes for each set $(-p_0,p_1,\cdots,p_n)$, with $p_0 = -(p_1+\cdots+p_n)$, where $\mathbf{E}_{p_0}, \mathbf{E}_{p_1},\cdots,\mathbf{E}_{p_n}$ appear in $\mathbf{E}_{-p}\cdot\mathbf{P}_p^{(n)}$, and finally that, within each such set, the factors of p_1, etc., p_n vanish separately. These factors are of course the terms

$$
\chi^{(n)\;T_{1\,j+1}}(-p_j\omega_I; p_0\omega_I,\cdots,p_{j-1}\omega_I,p_{j+1}\omega_I,\cdots,p_n\omega_I)
$$

$$-\chi^{(n)}(-p_0\omega_I; p_1\omega_I, \cdots, p_n\omega_I),$$

justifying the general criterion.

This general result appears in [10]; we hope that the physical meaning of these equations, as well as the assumptions we were led to carry out, have been clarified.

5 Conclusion and outlook

This paper explores a new route to obtain the propagation equation systems governing nonlinear interactions of light in matter. The aim is to obtain equations that do not depend on the particular effect, the particular involved material or the particular situation we want to study. This aim is partly fulfilled, especially when treating harmonic generation. For this, the introduction of the degree is of major importance. To our knowledge, no similar notion has been introduced in the literature.

The paper [3] is devoted to numerical results; the system (21) is chosen for its interesting effects (second and third harmonic generation, optical Kerr effect, depletion of the pump beam) and because it allows to make a power balance. Of course, only an experimental test could invalidate the theoretical as well as the numerical models. Though these experiments should not be difficult to carry out, we have to confess, without being to much ironic, that the result is highly uncertain. Indeed, we fear to fall into one of the two possibilities: either the incident power is too low for the nonlinear effects to be measurable, or, particularly because our incident beam is monochromatic, the medium will not be insensitive to light, so that thermal or mechanical effects should be included.

To conclude, the method exposed in this paper has to be generalized in several directions: to match experimental data, we have to deal with non-stationary media, to include non-harmonic generation (in order to study subharmonic generation, Raman scattering, etc.), and also to generalize our notion of the degree to the cases where the sources of the electromagnetic field oscillate at several frequencies (in order to study sum-harmonic generation, four-wave mixing, etc.), or even with a continuous spectrum (in order to deal with laser pulses).

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